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Evaluation of Ceramic Filters for High-Temperature/High-Pressure Fine Particulate Control

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16. ABSTRACT The report gives results of a study to analyze and evaluate ceramic membrane filters as a new, fine particulate (<3 micrometers) control concept for high-temperature (approx. 900 C), high-pressure processes. Several ceramic filters were identified as potantial candidates for fine particulate removal. There does not seem to be any inherent material limitation to high-temperature operation; however, no evidence of high-temperature filter application was found. The filters typically are 2-6 mm thick, cylindrical, and available with various pore sizes, increasing upward from 0.5 micrometer. These elements may be suitable for fine particulate control in hot gas streams. The most promising, although undeveloped, idea for a ceramic filter is to use ceramic honeycomb monoliths similar to those available for catalyst supports and heat exchangers. The walls of the monoliths are about 0.2-0.4 mm thick and of varying pore size and porosity. Geometric configurations are available which would force the gas to flow through the membrane walls. Pressure losses would be very small relative to those of standard ceramic filter elements. The application of ceramic monoliths to high-temperature fine particulate control appears very promising. It is strongly recommended that this concept be investigated further.

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FOREWORD

This document contains a review and evaluation of ceramic membrane filtration as a novel concept for the control of fine particulate. Several materials are identified as candidates for testing as high temperature, fine particulate filters and a test matrix is proposed.

The study was performed for the Environmental Protection Agency, Research Triangle Park, North Carolina. Dr. D. C. Drehmel was the EPA Task Officer. The Aerotherm Program Manager was Mr. Fred Moreno. Acting as Technical Advisor for the task was Dr. C. B. Moyer. The study was performed during the period December 1975 through June 1976.

TABLE OF CONTENTS

Section		Page
1	SUMMARY	1
2	INTRODUCTION	3
3	THEORETICAL DISCUSSION	4
	3.1 Filter Efficiency	5
	3.2 Pressure Drop	7
	3.3 Actual Filtration	10
4	CURRENT RESEARCH	13
5	LITERATURE AND MANUFACTURERS SURVEYS	14
	5.1 Thick Walled Elements	17
	5.2 Thin Walled Elements – Catalyst Monolith Supports	20
	5.3 Potential Cleaning Methods	20
6	PROPOSED TEST PROGRAM	31
7	PROCESS ECONOMICS	36
8	CONCLUSIONS AND RECOMMENDATIONS	38
	REFERENCES	39
	APPENDIX A – FILTER TEST PLAN	40

LIST OF ILLUSTRATIONS

Figure		Page
1	Collection efficiency versus particle size	8
2	Collection efficiency versus particle size	9
3	Pressure response in constant rate gas filtration	11
4	Multiple element filter assembly	18
5	Flow rate of air at standard conditions for a typical ceramic cylindrical element, wall thickness ~ 6.0 mm	19
6	Examples of 3M Company's ThermaComb corrugated ceramic	21
7	Structural shapes for 3M ThermaComb	22
8	Examples of Corning's Celcor cordierite monoliths	23
9	Examples of Dupont's Turvex honeycomb	24
10	Example of General Refractory Company's Versagrid ceramic honeycomb	25
11	Examples of Norton Company's silicon carbide Spectramic honeycomb	26
12	Pore size distribution for 3M Company's ThermaComb AlSiMag 795	27
13	Pressure drop across an 0.2 mm (0.008 in) AlSiMag 795 flat ceramic piece	28
14	Air flow through 0.2 mm (0.008 in) AlSiMag 795 flat ceramic piece	29
15	3M Crossflow Ceramic Monolith	32
16	3M Element Low Temperature Holder	33
17	3M Element and Holder Mounted Inside Pipe	34

LIST OF TABLES

Table		Page
1	Filter Operating Conditions	15
2	Survey Results	16
3	Comparison of Filter Element Costs for the Conditions in Table 1	37

SECTION 1

SUMMARY

The purpose of this study was to analyze and evaluate ceramic membrane filters as a new, fine (<3μm) particulate control concept for high temperature (~900°C), high pressure (HTHP) processes. The results of this effort are summarized in the following paragraphs.

The concept of membrane filters is not a new one. A review of the theory of operation, from the standpoint of both particle collection efficiency and fluid flow behavior, is presented here. The theory is the same as that for fiber filter operation. The complexity of the problem necessitates that many simplifying assumptions be made; consequently, theoretical calculations are not very reliable for predicting actual filter operation. However, these calculations can be very useful in explaining the qualititative behavior of a filter and in investigating the sensitivity of filter operation to changes in fluid parameters and filter material characteristics. Actual filter operation can be predicted only with the aid of sufficient experimental data at similar operating conditions.

Current research to develop ceramic membrane filters for use in HTHP processes is limited to an EPA sponsored project with Westinghouse and Horizon Research Inc. The purpose of this project is to develop a thin-walled (~0.6 mm) ceramic material with a pore size of approximately 0.5 μm. Success in producing a sample suitable for laboratory testing as a filter material has been limited.

Several ceramic filters were identified in this study as potential candidates for fine particulate removal. There does not seem to be any inherent material limitation to high temperature operation; however, no evidence of high temperature filter application was found. These filters are typically 2 to 6 mm thick, cylindrical in shape, and available with various pore size increasing upwards from 0.5 μm. It appears that these elements may be suitable for fine particulate control in hot gas streams.

The most promising, although undeveloped, idea for a ceramic filter is to use ceramic honeycomb monoliths similar to those available for catalyst supports and heat exchangers. The walls of the monoliths are approximately 0.2 to 0.4 mm thick and of varying pore size and porosity. Geometric configurations are available which would force the gas to flow through the membrane walls.

Pressure losses would be very small relative to those of standard ceramic filter elements. The application of ceramic monoliths to high temperature fine particulate control appears very promising, and it is strongly recommended that this concept be further investigated.

Three ceramic materials were selected for testing and were delivered to Westinghouse. The materials were a standard ceramic filter material (Selas), a ceramic monolith (3M) and a new material called ceramic FiberForm (FMI). A test matrix was prepared and is included as Appendix A.

A combined-cycle power plant using ceramic membrane filters appears to be cost competitive with other combined-cycle fine particulate removal systems. The ceramic monolith concept offers a substantial cost savings (perhaps a factor of 10) over other ceramic filter elements.

Ceramic filters appear to be both technically and economically feasible as fine particulate control devices in high temperature gas flows; however, suitable experimental data must be obtained in order to adequately evaluate them. Each type of filter should be tested under conditions substantially similar to the expected operating conditions. Data concerning the collection efficiency, pressure loss, duty cycle, filter life, and methods of cleaning should be obtained.

SECTION 2

INTRODUCTION

High temperature gas turbines used to generate electric power require gas streams virtually free of particulate matter. Gas streams from high temperature, high pressure coal processes, such as low Btu gasification and pressurized fluidized bed combustion, require considerable particulate removal. In order to maintain high thermal efficiency the particulate clean-up must be done at the high temperatures of the process. Coarse particles (>10μm) can be successfully removed by high temperature cyclones. Many new concepts for fine particulate control at elevated temperatures are presently being proposed. One such concept utilizes ceramic membrane filters.

Ceramic materials have been used extensively in high temperature environments. High melting temperature and mechanical strength, low thermal expansion, and resistance to chemical attack are some of their attractive features. These materials have found application as filters in medicine, biology, aerospace, and electronics where extremely clean gas and liquid streams are required. There are no obvious reasons why ceramic filters would not also be suitable for the high temperature, fine particulate control applications described here.

This report presents a review and evaluation of current, proposed and potential ceramic membrane particulate control technology. The theory of membrane filters is reviewed in Section 3. Section 4 is a brief summary of the current EPA-Westinghouse ceramic membrane filter program, which is currently the only developmental research program concerning ceramic filters for high temperature applications. Section 5 presents the results of the literature and manufacturers surveys. Three materials, proposed for testing at Westinghouse, and a test plan are discussed in Section 6. The economics of ceramic filters are briefly discussed in Section 7, and the conclusion and recommendations presented in Section 8.

SECTION 3

THEORETICAL DISCUSSION

Filter materials are usually classified as one of two types, membrane filters or fiber filters. In a two dimensional representation, membrane filters may be thought of as a uniform solid phase interrupted by a fluid phase (holes), and are generally characterized by low porosity (void fraction). A fiber filter may be thought of as a uniform fluid phase (holes) interrupted by a solid phase (fibers) and is characterized by high porosity. In an oversimplified view, membrane filters work because the membrane pore diameter is smaller than that of the particles to be collected; the particles are thus prevented from passing through the membrane. On the other hand, fiber filters operate most effectively after an initial "cake" (layer) of particles has been deposited; thus, the cake serves as the filter. In either case the theoretical analysis of the filter operation is the same and follows directly from the theory of flow through porous media. The analysis results in the development of two measures of filter performance – particle collection efficiency and fluid pressure drop across the filter.

Usually, the theoretical development of filter collection efficiency has been approached microscopically from a very detailed analysis of small scale events inside the filter. Important parameters are the particle size, filter pore size, fluid Reynolds number, and diffusivity. Each collection mechanism is studied separately and an expression for the collection efficiency derived. The individual expressions are then combined to yield an estimate of the total collection efficiency of the filter. These expressions are usually derived for a single size particle and well-defined flow conditions. Unfortunately, the procedure is quite complicated, necessitating the use of many simplifying assumptions which can lead to considerable inaccuracy.

The analysis of fluid pressure drop, on the other hand, is approached macroscopically. The system is treated as a continuum and the suitably modified equations of motion are applied. A filter porosity (ε) is introduced in the continuity equation and a filter permeability (κ) modifies the momentum conservation equation. Porosity is a straightforward measure of filter void fraction and may be easily calculated from density measurements. The permeability, however, is a measure of the fluid drag introduced by the filter and cannot be accurately predicted at present. It thus becomes an empirical constant.

Expressions for the efficiency and the pressure drop (from Darcy's law) of a filter are developed for highly idealized systems in the following sections. This is followed by a discussion of the possible effects of some real system complications.

3.1 FILTER EFFICIENCY

Particle collection by filtration results from the action of three basic mechanisms:

- Diffusion of particles to solid surfaces
- Impaction of particles on solid surfaces due to deviation of the particle motion from the streamlines of fluid motion
- Interception of particles by solid surfaces due to particle motion along streamlines.

Clearly, the last two mechanisms depend on the details of the fluid flow adjacent to the pore surfaces. Some authors describe a fourth mechanism – sieving, in which a particle with a radius greater than the largest pore radius is "strained" out of the fluid. However, this is just a special case of impaction.

A detailed analysis of each collection mechanism is quite tedious and complicated, and will not be presented here. The resulting equations, as given by Davies (Reference 1), are presented instead. The analysis assumes that particles are chemically inert, uncharged, solid spheres of identical size (mono-disperse). In addition, particles are assumed to be at infinite dilution so that particle – particle interactions and pore clogging may be neglected. Thus the porosity and permeability can be considered constants. The resulting expressions for filter efficiency include the pore radius (R), the mean fluid velocity in each pore (v) and the average length of the pore (h) as parameters. These quantities are determined from measurements of the porosity, the average number of pores per unit area, and the fluid flowrate.

The efficiency due to diffusion (E_D) has been given by Spurny & Pich (References 2,3,4) as:

$$E_D = 1 - 0.81904 \exp(-3.6568\, N_D) - 0.09752 \exp(-22.3045\, N_D) - 0.03248 \exp(-56.95\, N_D) - 0.0157 \exp(-107.6\, N_D), \quad (1)$$

where $N_D = h\,\nabla/R^2 v$

$\nabla = kTF/6\pi a\mu$ = Diffusion coefficient

F = Cunningham slip factor

k = Boltzman's constant

a = particle radius

μ = fluid viscosity.

The expression is valid for pore radii smaller than 10 percent of the filter thickness.

The efficiency due to inertial deposition (E_I) is given by Pich (References 5,6) as

$$E_I = 1 - (\alpha + \beta)^2/(\alpha + 1)^2, \tag{2}$$

where $\alpha = 1/\left(\sqrt{\pi/2\sqrt{3}\,\varepsilon} - 1\right)$

ε = porosity

$\beta = 1 - 2S\sqrt{\alpha} + 2S^2\alpha\,(1 - \exp(-1/S\sqrt{\alpha}))$

$S = mFv/6\pi a\mu R$ = Stokes number of particle

m = mass of particle.

This is an unproven expression in that many unsupported approximations were used in its derivation; however, it is possibly the most accurate expression presently available.

The efficiency due to interception (E_R) is determined geometrically as the ratio of the interception area of the pore ($\pi R^2 - \pi(R - a)^2$) to the total area of the pore (πR^2). Thus,

$$E_R = \frac{a}{R}\left(2 - \frac{a}{R}\right). \tag{3}$$

The overall collection efficiency (E) is then given by:

$$E = E_I + (1 - E_I)(E_D + \gamma E_R), \tag{4}$$

where $\gamma = 0.6^{(1\,+\,a/R)}$.

When this number exceeds unity the efficiency is assumed to be 100 percent. The overall collection efficiency is seen to be a function of particle size, pore size, porosity, and particle density. Fluid properties of importance are the viscosity, the Cunningham slip factor, the flowrate and the fluid mean velocity in the pore. The effects of pore size distribution on collection efficiency have been found to be quite significant (Reference 7); however, these effects are not included here.

Figure 1 shows the predicted overall collection efficiency as a function of particle size with superficial fluid velocity as a parameter. These results are for a membrane filter 0.25 mm thick with an ε of 0.3 and uniform pore radius of 5 microns. As expected, for larger particles (>0.5μm radius) collection efficiency increases with increasing particle size. However, note that for very small particles the predicted collection efficiency increases with decreasing particle radius. This results from enhanced diffusion of these particles to the collector surface. Figure 2 shows overall collection efficiency as a function of particle size with filter pore radius as a parameter.

3.2 PRESSURE DROP

The flow of a continuum fluid through a porous medium is described by the continuity and conservation of momentum equations. For a chemically inert, electrically neutral, homogeneous Newtonian fluid these equations are:

$$\varepsilon \frac{\partial \rho}{\partial t} = -\vec{\nabla} \cdot \rho \vec{V}_0 \tag{5}$$

$$\vec{V}_0 = -\frac{\kappa}{\mu}(\vec{\nabla} P), \tag{6}$$

where V_0 is the superficial velocity of the fluid at the surface of the porous medium, ε is the porosity, κ is the permeability and μ is the gas viscosity. Equation (6) is commonly called Darcy's law. For steady flow of a clean, ideal gas of constant viscosity these equations may be solved for a variety of geometries. The most common form of Darcy's law for flow through a planar filter of thickness L is:

$$V_0 = \frac{Q}{A} = \frac{\kappa \Delta P}{\mu L}, \tag{7}$$

where Q is the volumetric flowrate and A is the surface area and ΔP has been assumed to be small compared to P. This equation is often used to calculate the permeability of a material, since permeability is not derivable from other basic considerations. For a system of two planar filters of thickness L_1 and L_2

$$\Delta P = \Delta P_1 + \Delta P_2 = \frac{Q\mu L}{A\kappa}, \tag{8}$$

where $L = L_1 + L_2$ and

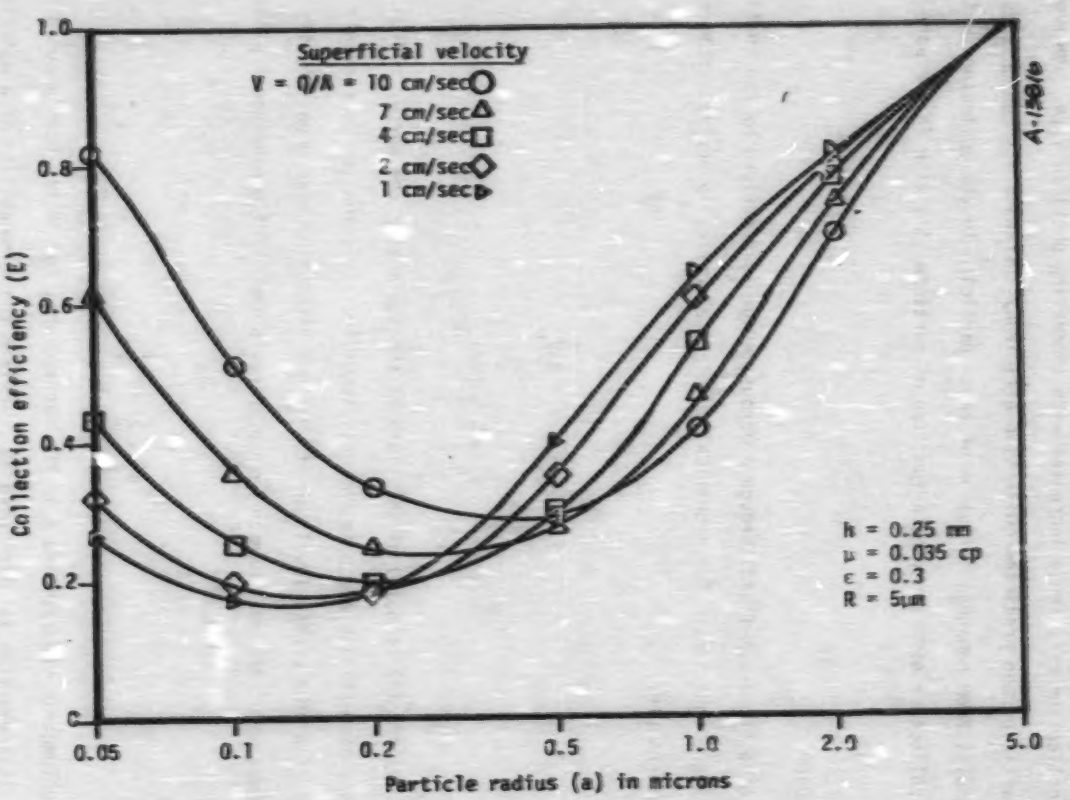


Figure 1. Collection efficiency versus particle size.

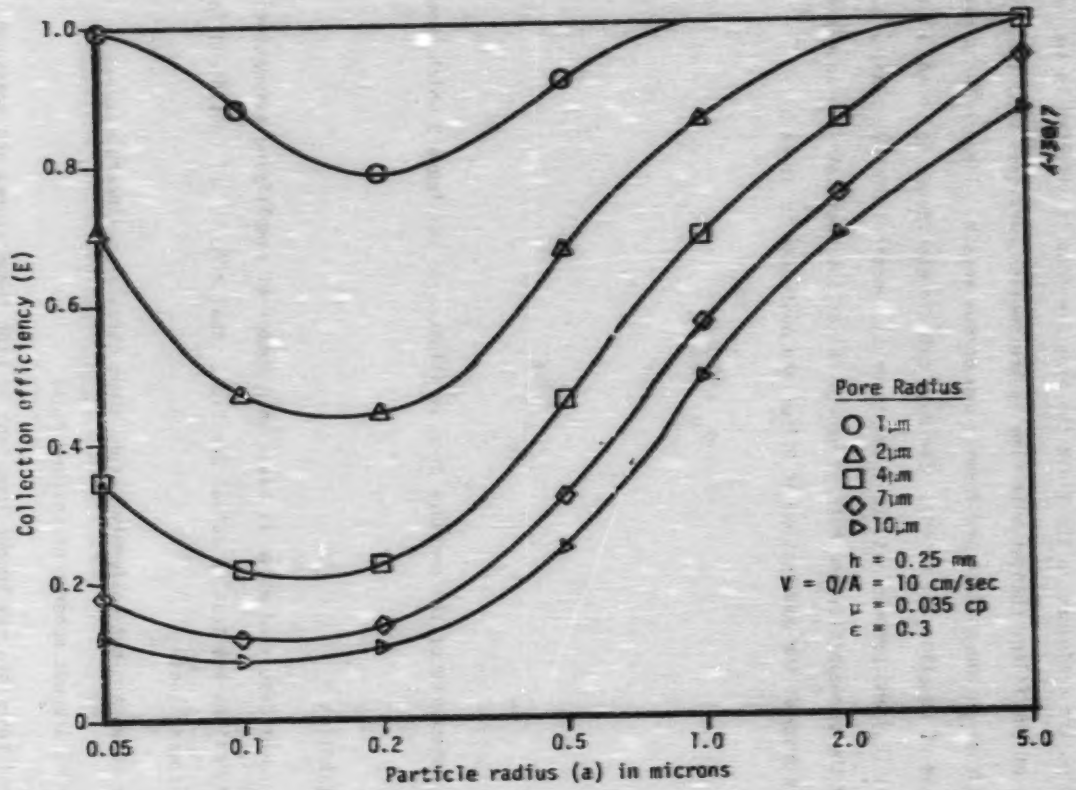


Figure 2. Collection efficiency versus particle size.

$$\kappa = L\kappa_1\kappa_2/(L_2\kappa_1 + L_1\kappa_2). \tag{9}$$

This form of Darcy's law is useful for determining the permeability of a cake-filter system with clean air. Similar results can be obtained for a cylindrical system.

If particles are contained in the fluid the pressure drop is affected in two ways. First the permeability of the filter changes with time as the smaller particles plug the pores. Second, a dust cake builds up on the surface and forms a new porous medium. These effects can be quantified by appropriate use of Equation (8) or (9). The dust cake is considered to be one of the filters and its thickness (L_2) and permeability (κ_2) are taken as functions of time. After a short initial transient period the permeability and rate of growth (k) of the dust cake are approximately constant; thus the pressure drop of the combination of filter and dust cake is given by:

$$\Delta P = \Delta P_0 + \frac{Qukt}{A\kappa_2}, \tag{10}$$

where

$$\Delta P_0 = \frac{Qu L_1}{A\kappa_1}. \tag{11}$$

This linear time dependence of the pressure drop is illustrated in Figure 3 which shows data for a fiber filter.

3.3 ACTUAL FILTRATION

The development described above is based on a number of simplifying assumptions. Few, if any, of these assumptions are true in actual practice. Real effects not considered in the above theoretical development include:

- Filter pores are not uniformly sized, right circular cylinders
- Particles are not spheres of uniform radii
- Chemical reactions may become important at high temperatures and pressures
- Corrosion of filter material may be important at high temperatures or in adverse environments
- Pressure fluctuations may cause changes in dust cake properties
- The effects of reentrainment may be significant.

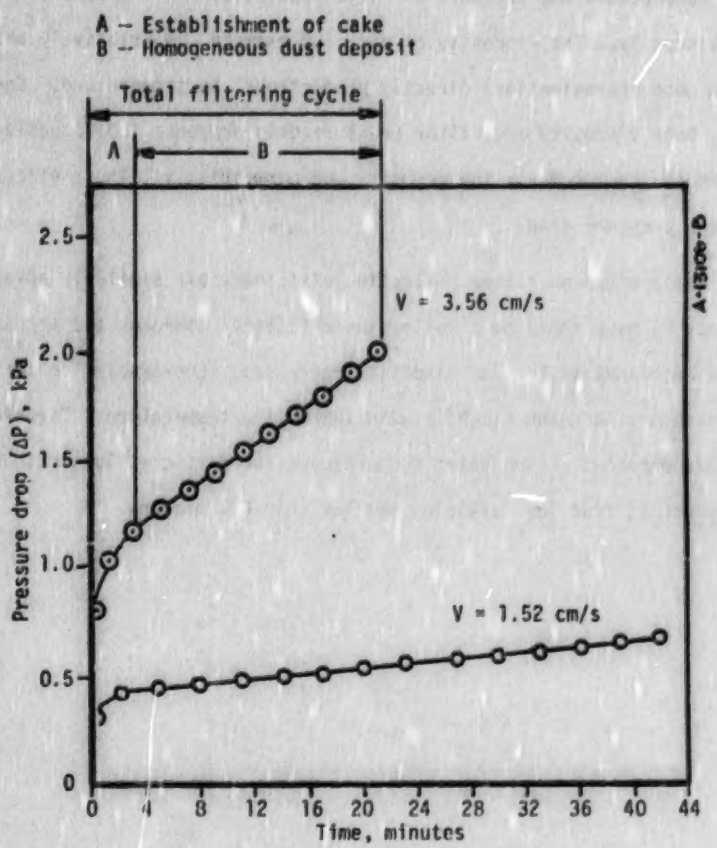


Figure 3. Pressure response in constant rate gas filtration.

Consequently, the expressions for efficiency and pressure drop should be taken as approximations, useful only to illustrate functional relationships of important system parameters.

The effects of temperature and pressure on filter operation are introduced largely through changes in the fluid viscosity. The viscosity of air, for example, is relatively unaffected by pressure, but is (to a good approximation) directly proportional to temperature. Consequently, as temperature increases, both viscosity and filter pressure drop increase (cf. Equation (10)). In addition, thermal expansion can decrease the porosity and permeability. These effects also give rise to increased filter pressure drop.

The effects of temperature on filter collection efficiency are similarly adverse. Strauss and Lancaster (Reference 8) have shown that collection efficiency (through the mechanisms of impaction and interception) decreases as the gas viscosity increases. Conversely, collection efficiency due to particle diffusion may increase slightly with increasing temperature. Therefore lower collection efficiencies are expected at increased temperatures for particles larger than 0.2 microns in diameter. The converse is true for particles smaller than 0.2 microns.

SECTION 4

CURRENT RESEARCH

Westinghouse Research Laboratories is currently performing the only large scale effort directed toward applying ceramic membrane filters to particulate removal from hot gas streams. This study has potential application to cleaning high temperature fuel gas from coal gasifiers, and is being funded by the EPA (Reference 9). The Westinghouse effort seeks to develop thin (0.25 mm) cylindrical membrane filter elements with small, uniform pore size (0.5μm). These filter elements are then to be tested and evaluated as a technique for collecting sub-micron particulate from a high temperature (>1500°F) fuel gas stream.

To date progress in obtaining appropriate planar samples of the filter material for performance testing has been limited. Fabrication problems have included:

- Difficulty in laying down a planar membrane free of oxide irregularities, which can become large holes when the membrane is etched
- Difficulty in converting the initially deposited γ-alumina to α-alumina without warping the filter disk (warped disks break when installed in filter holders for performance testing)
- Difficulty in uniformly enlarging the pores throughout the membrane to produce a filter with acceptable pore size.

As a result of these problems, few data for either clean gas flow characteristics of particulate removal performance are available. Recently Horizon Research has fabricated a suitable ceramic membrane and support structure, and has performed several successful preliminary performance tests. However, it appears that development of this material will be curtailed due to continued difficulties in preparation of suitable samples.

SECTION 5

LITERATURE AND MANUFACTURERS SURVEYS

There is a paucity of data available on the use of ceramic filters to collect particulate from hot gas streams. A limited survey of filtration literature and of ceramic manufacturers/suppliers was therefore undertaken to determine:

- The available types of ceramic filter materials
- The use of ceramic filters in current filtering applications
- The types of ceramic filter materials which may be suitable for collecting sub-micron particulate from a high temperature gas stream.

The survey was accomplished in four steps:

- Review of existing filtration literature
- Telephone contact with ceramic filter suppliers
- Telephone contact with ceramic manufacturers
- Telephone contact with monolithic catalyst support manufacturers.

The literature survey revealed that sub-micron particle ceramic filters are in use in medical aerospace, electronic and biological applications for both gases and liquids. However, no discussion of the operation of these filters at high temperatures was found. This appeared to be more a case of lack of present applications rather than ceramic material limitations.

These findings prompted the telephone survey discussed below. The purpose of the survey was to contact a representative sample of ceramic manufacturers and suppliers, and to determine if capabilities exist to develop a suitable rigid ceramic filter for application to hot gas clean-up. As a guide, a representative set of operating conditions was established (see Table 1). The conditions in Table 1 approximate those from a low BTU coal gasifier downstream of the cyclones. The key parameters are the temperature and the particle size limits. A summary of the relevant telephone contacts is given in Table 2.

TABLE 1. FILTER OPERATING CONDITIONS

Temperature	1100 to 1400°K	(1500 to 2000°F)
Pressure	2000 kPa	(300 psia)
Gas Flow Rate	200 m^3/min	(7000 SCFM)
Gas Composition (% Vol)	H_2	21
	CO	14
	H_2O	13
	CO_2	11
	N_2	38
	CH_4	2.5
	H_2S	0.5
	COS	30 ppm
	NH_3	600 ppm
	tars, heavy HC, others	trace
Molecular Weight	23	
Density	4.8 kg/m^3	(0.3 lbm/ACF)
Heat Capacity	2510 J/kg°C	(0.6 Btu/lbm°F)
Viscosity	3.5×10^{-5} N-s/m^2	(0.035 centipoise)
Particulate Loading (upstream of filter)	1.144×10^{-3} $\frac{kg}{m^3}$	(0.5 grain/SCF)
Distribution (by weight)	100% < 30μm	
	80% < 10μm	
	50% < 4μm	
	15% < 1μm	
Particulate Loading (downstream of filter)	1.144×10^{-5} $\frac{kg}{m^3}$	(0.005 grain/SCF)
Distribution (by weight)	100% < 6μm	
	98% < 4μm	
	95% < 2μm	
	80% < 1μm	

TABLE 2. SURVEY RESULTS

Company	Code*	Comments
1. Babcock & Wilcox Co., Refractories Div. Augusta, Georgia	c	Did not think they had any materials that would work for the application.
2. Coors Porcelain Co. Golden, Colorado	c,f	Several small scale filters for this particle size. No high temperature filter experience. Samples sent to Aerotherm. Nominal wall thickness ~6 mm (1/4").
3. Corning Glass Co.	f,s	Nothing for this application. Celcor cordierite monoliths a possibility.
4. Dollinger Corp. Rochester, New York	f	Some filter elements that would work for this application. Relatively thick wall, large pressure drop (~100 kPa (1 atm)). Elements available for testing.
5. General Refractories Bala Cynwyd, Pa.	c,s	Nothing for this application. Versagrid cordierite honeycomb is a possibility; however geometrical constraints a problem.
6. Horizons, Inc. Cleveland, Ohio	c	This membrane (~0.25 mm thick) being developed for testing in this application under Westinghouse – EPA program.
7. 3-M Company Saint Paul, Minn.	c,s	AlSiMag 795 (cordierite) honeycomb structure should work for this application. Elements available for testing. Low pressure drop. High surface area to volume ratio. Good process control over pore size, porosity and membrane thickness. Samples sent to Aerotherm.
8. Norton Company Worcester, Mass.	c,s	Two materials suitable for thick walled filters. Also Spectramic honeycomb is a possibility.
9. Selas Flotronics Springhouse, Pa.	f	Micro-porous porcelain element would be suitable. Pressure drop ~50 kPa. Sample sent to Aerotherm.
10. Wisconsin Porcelain Co. Sun Prairie, Wisconsin	c,f	Thick walled filter elements similar to 4 and 9. No membranes.

*c - ceramic manufacturer
f - filter supplier
s - catalyst support manufacturer

All contacts were asked to supply information pertaining to:

- High temperature operation
- High flowrate operation
- Cleaning procedures
- Cost
- Novel applications and design ideas.

In addition, all contacts were asked if they were supplying anyone with ceramic elements suitable for hot gas cleaning, if they were developing any materials suitable for this application, and what if any, material limitations should be considered for this application.

In general, two types of ceramic materials suitable for this application were identified. These are discussed separately in the following sections, and a brief discussion of possible filter element cleaning methods is also given.

5.1 THICK WALLED ELEMENTS

The type of filter element generally available from ceramic manufacturers and filter suppliers is referred to as a "thick walled element". It is usually cylindrical and has a wall thickness of 4 to 6 mm which is "thick" in comparison to the Westinghouse/Horizons membrane and ceramic monoliths (wall thickness ~0.25 mm). For large flowrate applications, the elements (typically 50-100 mm in diameter and 250-300 mm in length) would be grouped in an assembly similar to the one shown in Figure 4. The wall thickness of the element is somewhat limited by the ceramic forming process; however, a more significant limitation is mechanical strength. The structural strength of the wall decreases with decreasing wall thickness; hence, thin walls in the cyclindrical geometry would probably be too fragile for heavy duty use.

Thick walled elements have several inherent disadvantages. First, the pressure drop across an element increases directly with the wall thickness (see Equation (11)). Typical pressure losses for a 6 mm thick filter of this type are shown in Figure 5 with pore size as a parameter. Values on the order of 35 to 70 kPa (5 to 10 psi) may be expected for filters of this type. A second problem with thick walled elements is the possible occurrence of thermal stress and spalling when the element is subjected to temperature cycling. This requires considerable care in start-up and shut-down procedures. Finally, material costs for thick walled elements are greater than for thin walled elements. This is discussed in more detail in Section 6.

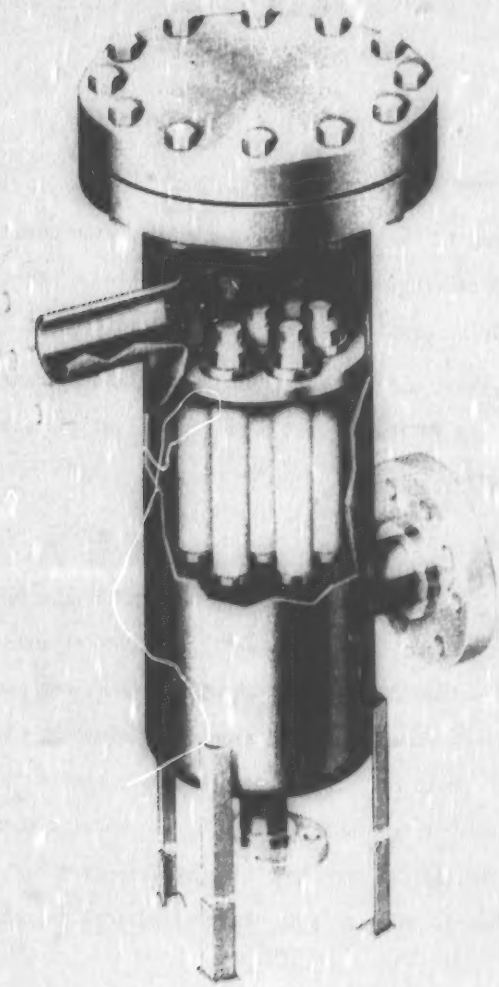


Figure 4. Multiple element filter assembly.

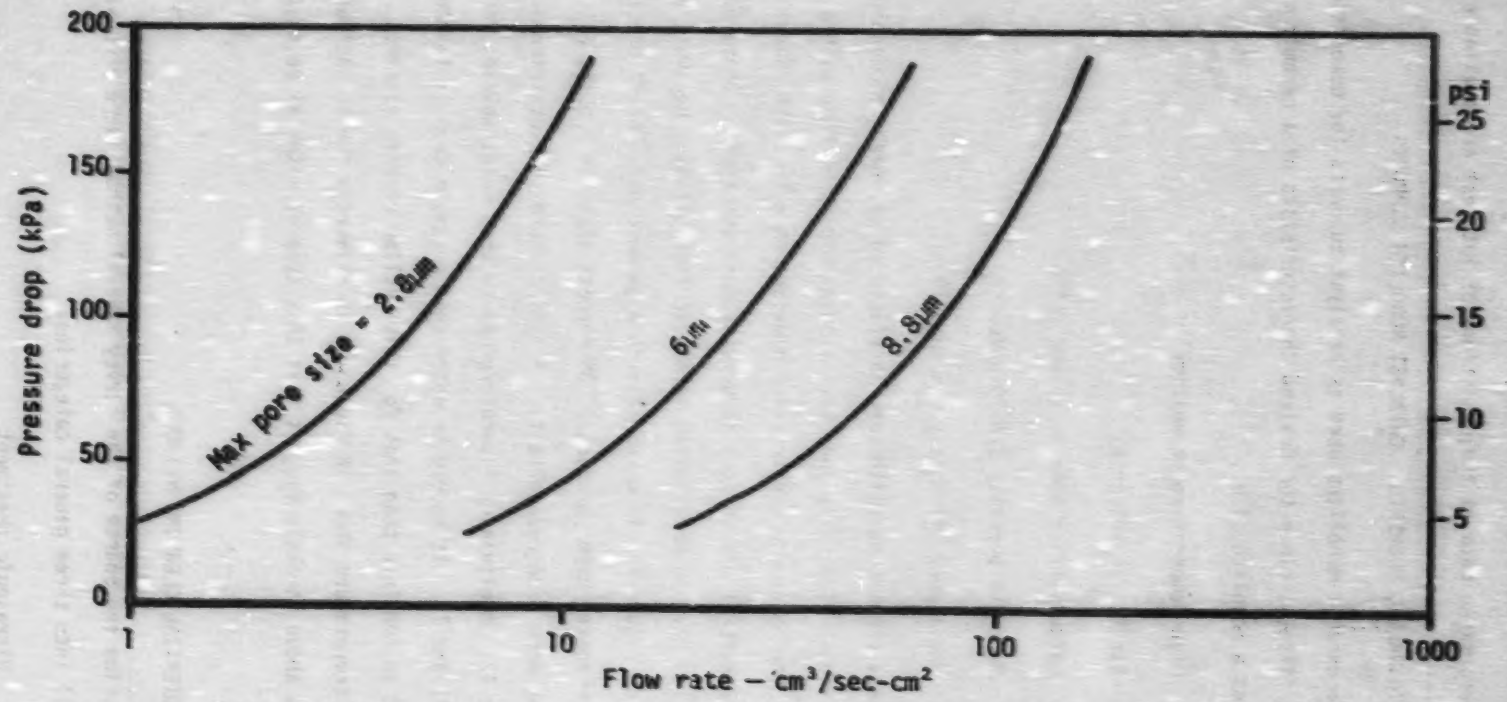


Figure 5. Flow rate of air at standard conditions for a typical ceramic cylindrical element, wall thickness ~ 6.0 mm.

The impact of the disadvantages described above will have to be assessed in light of other ceramic devices and particulate removal methods. An adequate assessment can be made only if operational performance data at the conditions of interest are available.

5.2 THIN WALLED ELEMENTS – CATALYST MONOLITH SUPPORTS

The ceramic monoliths used as catalyst supports for combustion processes were considered as potential filters, since the desired characteristics of a ceramic membrane filter are very similar to those of a monolith, i.e.,

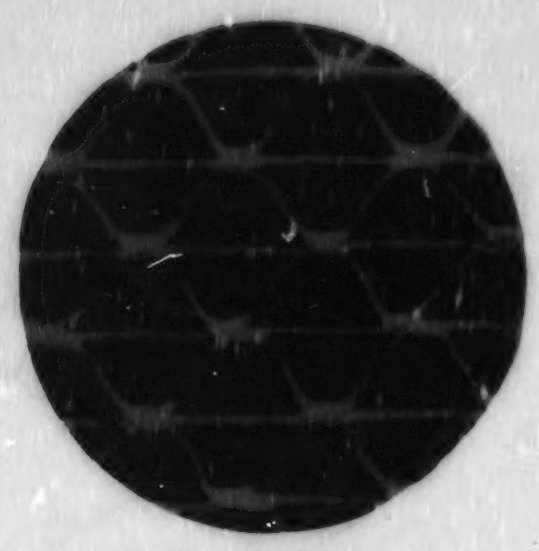
- High temperature capability
- Structural integrity
- Large surface area to volume ratio
- Moderate porosity (25-30 percent)
- Small pore diameter (less than 10μm).

These ceramic monoliths are manufactured by several companies (see Table 2) in a variety of configurations. Typically they have a honeycomb appearance with a wall thickness of approximately 0.25 mm. The honeycomb construction provides excellent structural strength. Figures 6 through 11 illustrate some of the various structural configurations. Those monoliths manufactured by an extrusion process have rather limited geometric flexibility. However, some processes (e.g., 3M Company as shown in Figure 7) allow very flexible geometries. The 3M process is similar to that used in making corregated paper products. 3M reports that the process allows independent control over wall thickness, pore size and porosity. A typical pore size distribution for one of 3M's products is shown in Figure 12. Preliminary flowrate/pressure drop relationships for the 3M material are shown in Figures 13 and 14. It should be noted that pressure drop is in the 1 to 10 kPa range which is a factor of 10 to 100 less than that for thick walled ceramic elements. The decrease in flowrate with increasing temperature shown in Figure 14 is primarily due to increases in gas viscosity. The advantag discussed above strongly suggest that this type of material be tested for use as a fine particulate filter.

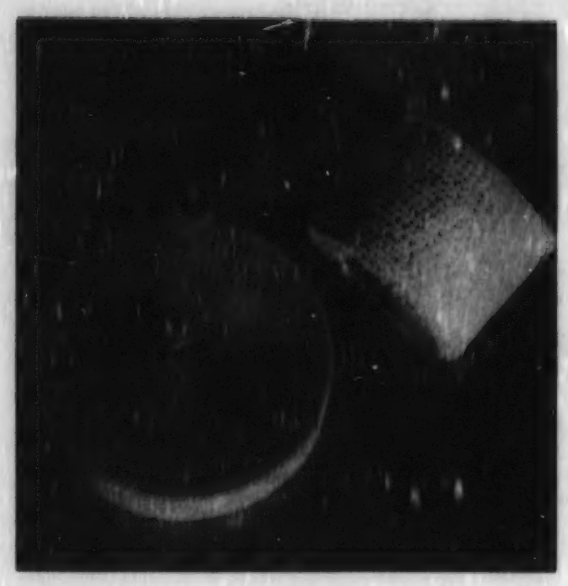
5.3 POTENTIAL CLEANING METHODS

During the course of the survey several methods were proposed for cleaning the filter elements These fell into three general categories:

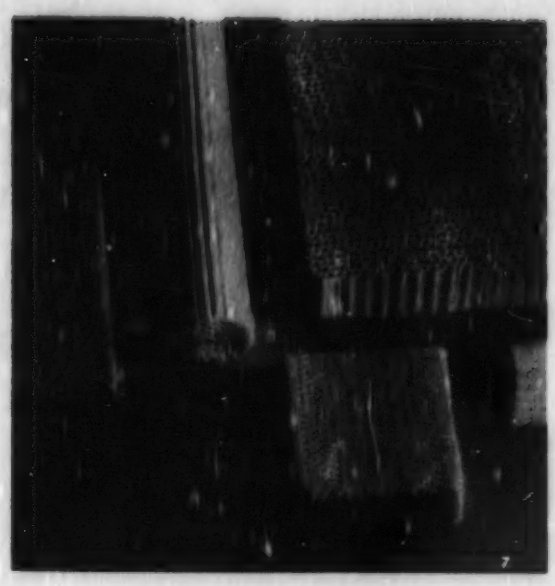
- Ultrasonic treatment
- Washing
- Back flow



a. 8x enlargment



b. Rolled structures



c. Rolled and stacked structures

Figure 6. Examples of 3M Company's ThermaComb corrugated ceramic.

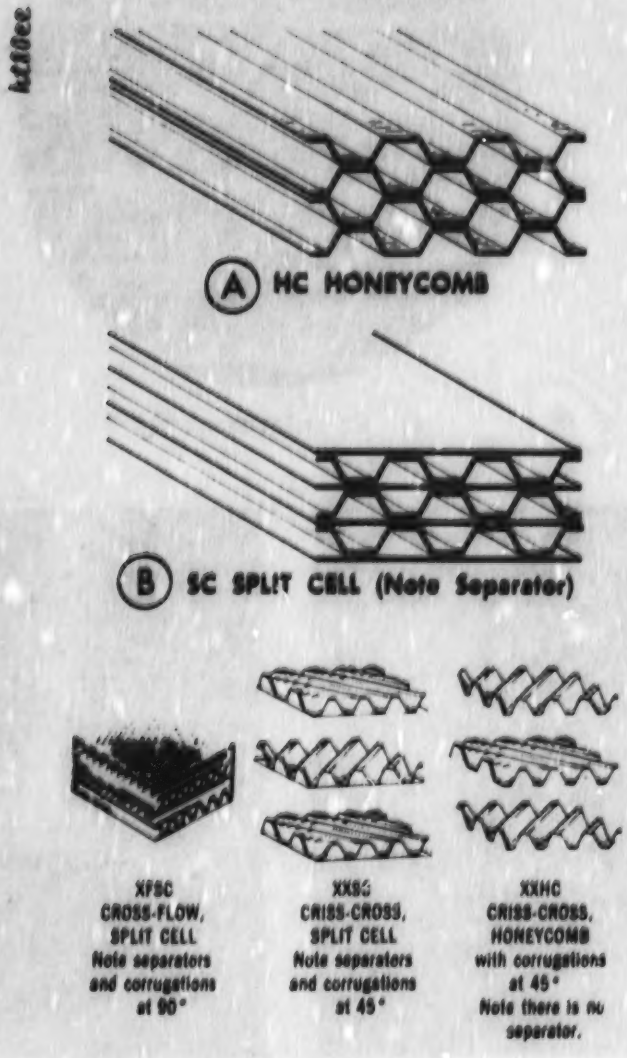
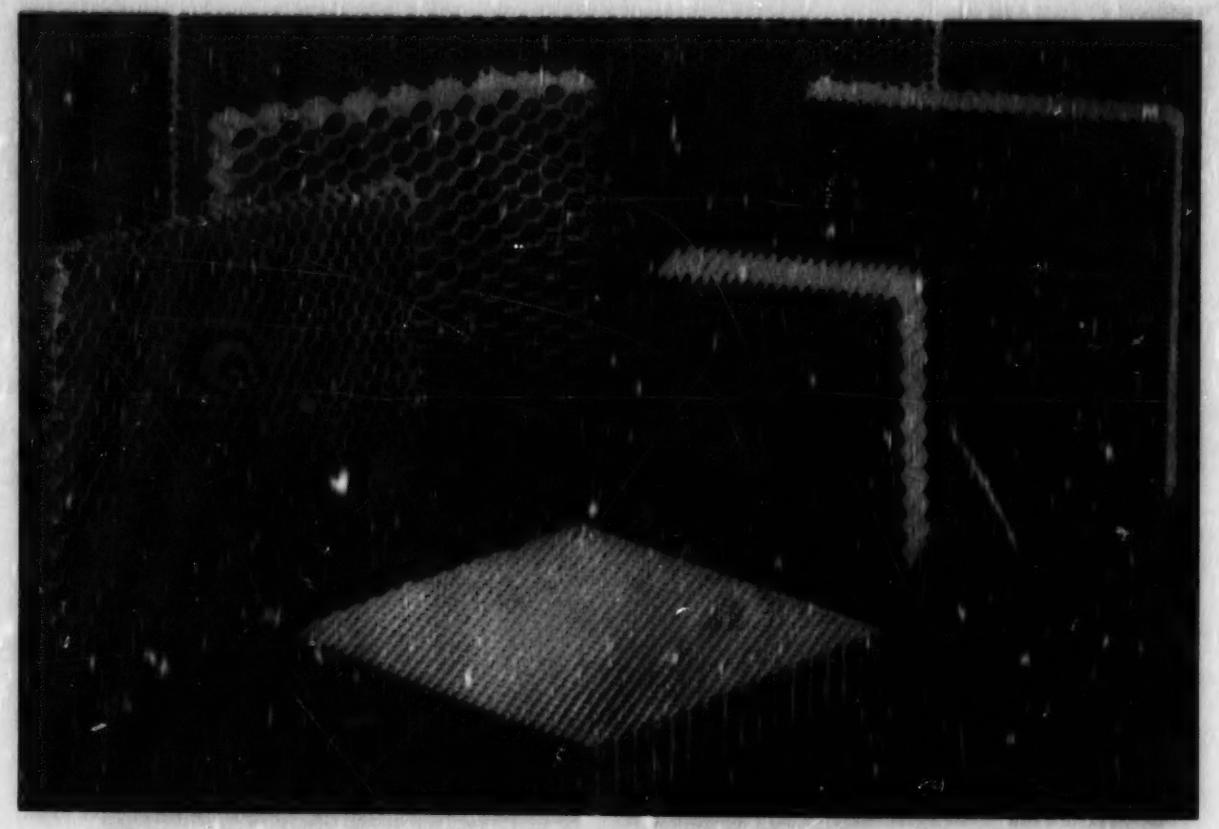
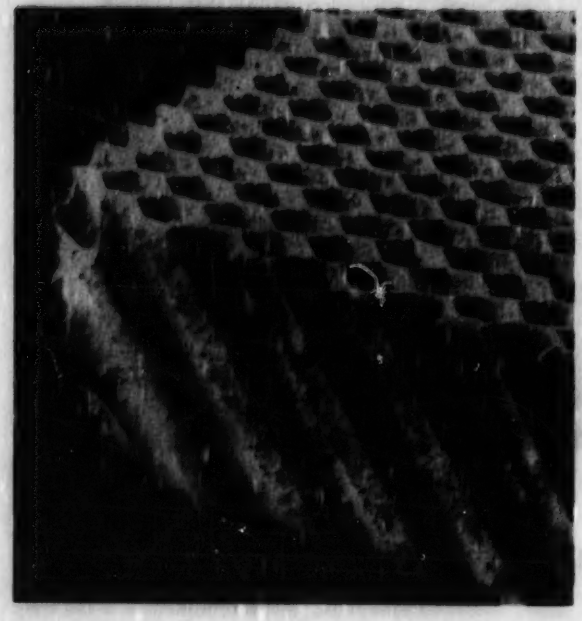


Figure 7. Structural shapes for 3M ThermaComb.

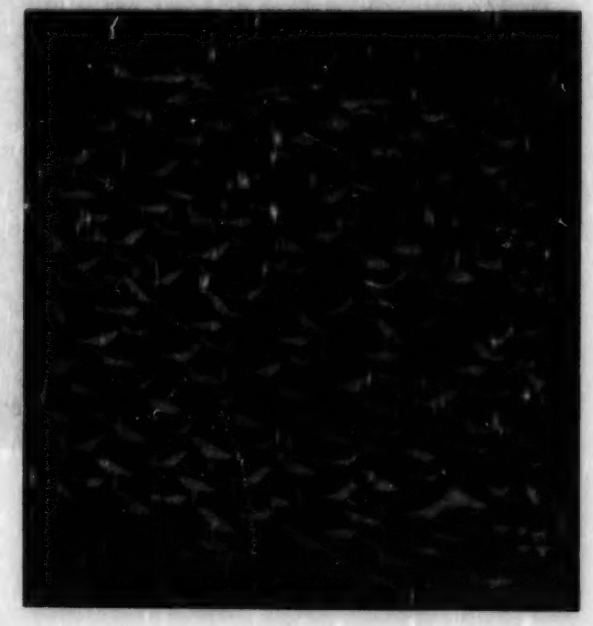
Figure 8. Examples of Corning's Celcor cordierite monoliths.



a. Straight honeycomb



b. Slant cell honeycomb



c. Crossflow honeycomb

Figure 9. Examples of Dupont's Turvex honeycomb.

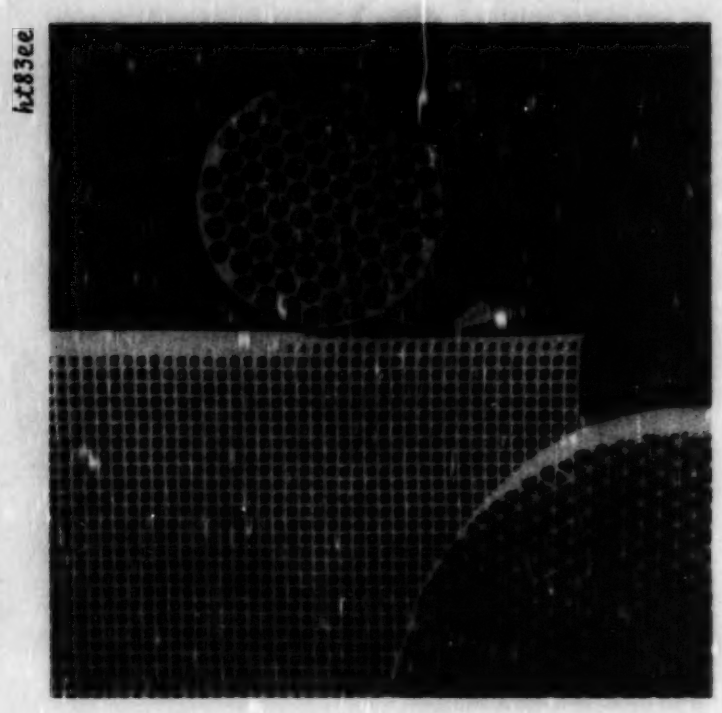


Figure 10. Example of General Refractory Company's Versagrid ceramic honeycomb.

Figure 11. Examples of Norton Company's silicon carbide Spectramic honeycomb.

Figure 12. Pore size distribution for 3M Company's ThermaComb AlSiMag 795.

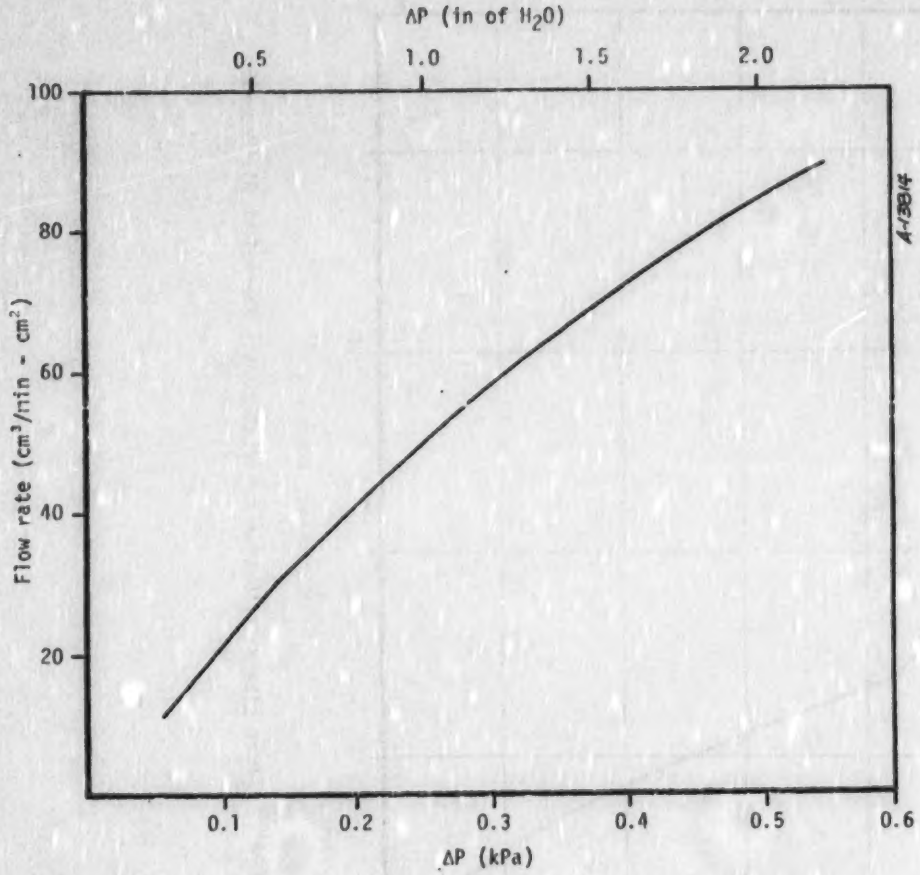


Figure 13. Pressure drop across an 0.2 mm (0.008 in) AlSiMag 795 flat ceramic piece.

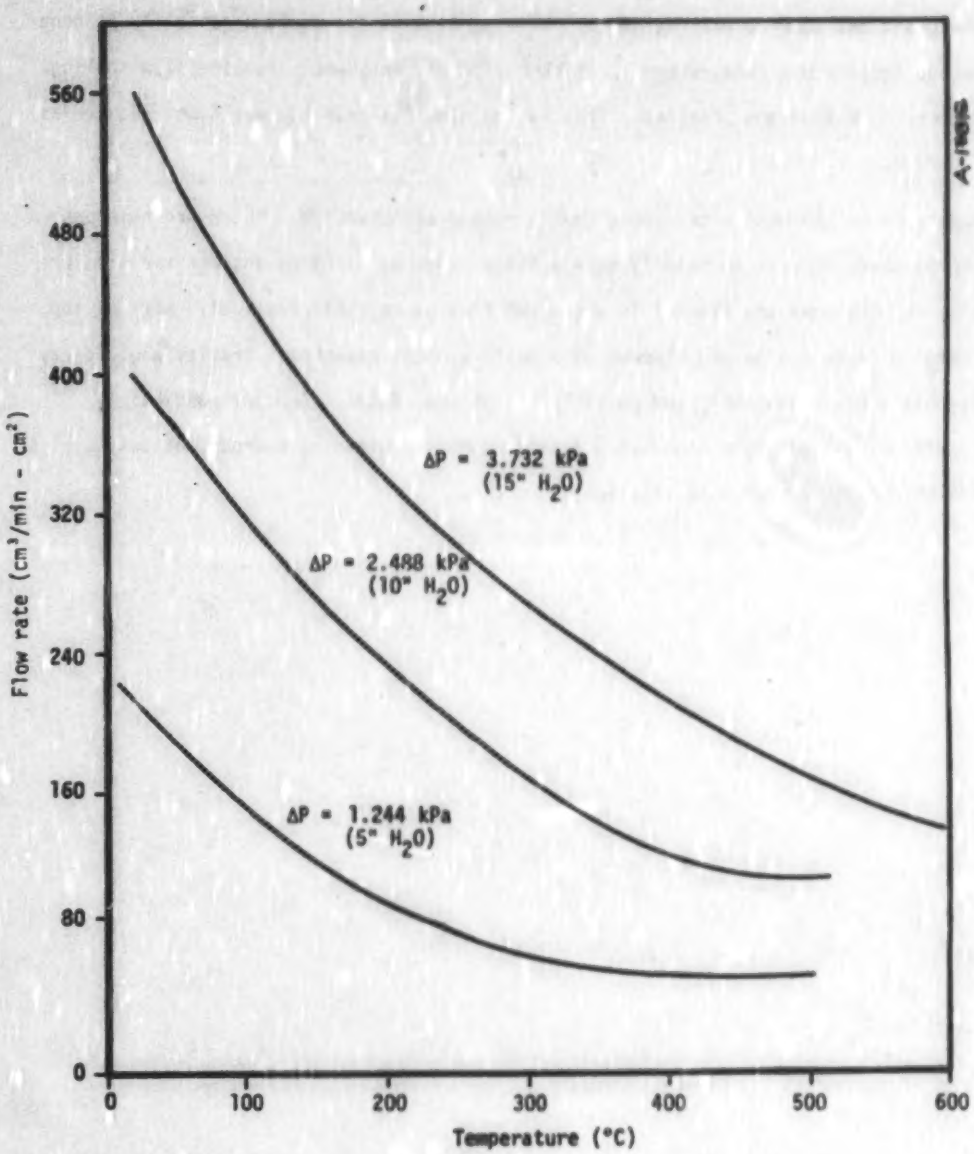


Figure 14. Air flow through 0.2 mm (0.008 in) AlSiMag 795 flat ceramic piece.

No specific information was obtained concerning either possible implementation methods or possible treatment efficiencies of sonic cleaning although it was generally thought that the application of ultrasonic energy would aid and speed a washing cycle. Washing of a filter would necessarily be done off-line due to time and temperature requirements. Thick walled elements would require slow cooling to prevent thermal stress, cracking and spalling. Thin walled elements probably would not require as lengthy a cool-down period.

Back flow appears to be the most promising cleaning method at this time. There are numerous ways this can be accomplished. One is to briefly take a filter element off-line and use the pressure in the filter assembly to blow down the element in a reverse flow pulse. Alternatively, part of the clean gas could be used to reverse flow one element of a multi-element assembly. Gravity would carry the particles to the bottom of the assembly and possibly out of the vessel. Another possibility is the use of a pressure pulse similar to baghouse cleaning methods. Cleaning methods and duty cycles still require considerable experimental investigation.

SECTION 6

PROPOSED TEST PROGRAM

At the conclusion of the work described in Sections 4 and 5 it was evident that the only way to realistically evaluate high temperature, high pressure ceramic filters would be to test the available materials at the conditions of interest. Three materials were identified for testing, and a test matrix was prepared. The materials and the test matrix are discussed below.

A standard ceramic filter manufactured by Selas Flotronics was selected as representative of the "thick walled" elements described in Section 5.1. This material has been tested and used in low temperature environments; however, there are no data available on the high temperature ($T > 200°C$) performance of this material. There is a relatively large pressure drop (50 - 100 kPa) across the element. It is expected that the pressure drop will increase at higher temperatures.

A cross-flow ceramic monolith manufactured by 3M was selected to represent the ceramic monolith elements described in Section 5.2. The primary reason for this choice was that this was the only monolith available in a cross-flow geometry. Preliminary data for the pressure drop-flowrate-temperature relationships were available from 3M (Figures 13 and 14). However, there are no data on the filtration characteristics of this material. Several samples of this material were provided by 3M. These samples and a low temperature element holder were delivered to Westinghouse for testing. The split cell cross-flow construction is shown in Figure 3. Sketches of the material and holder are shown in Figures 15, 16 and 17.

A third material, which is significantly different from the two described above, was also identified. This material, called alumina FiberForm, is manufactured by Fiber Materials, Inc. It is made of alumina-silica fibers bonded with a high purity alumina binder and produced using vacuum slurry molding techniques. FiberForm has a useful temperature limit of about 1450°C (2600°F). It is a very high porosity, low density material and has a texture somewhat like styrofoam. This material has never been tested as a filter; however, its pore size, porosity and temperature limits make it an attractive candidate. Samples of FiberForm have been delivered to Westinghouse.

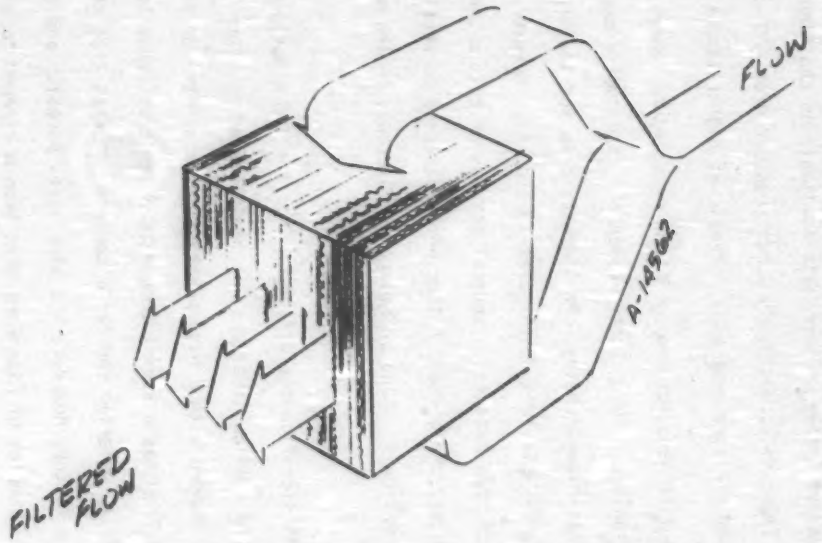


Figure 15. 3M crossflow ceramic monolith.

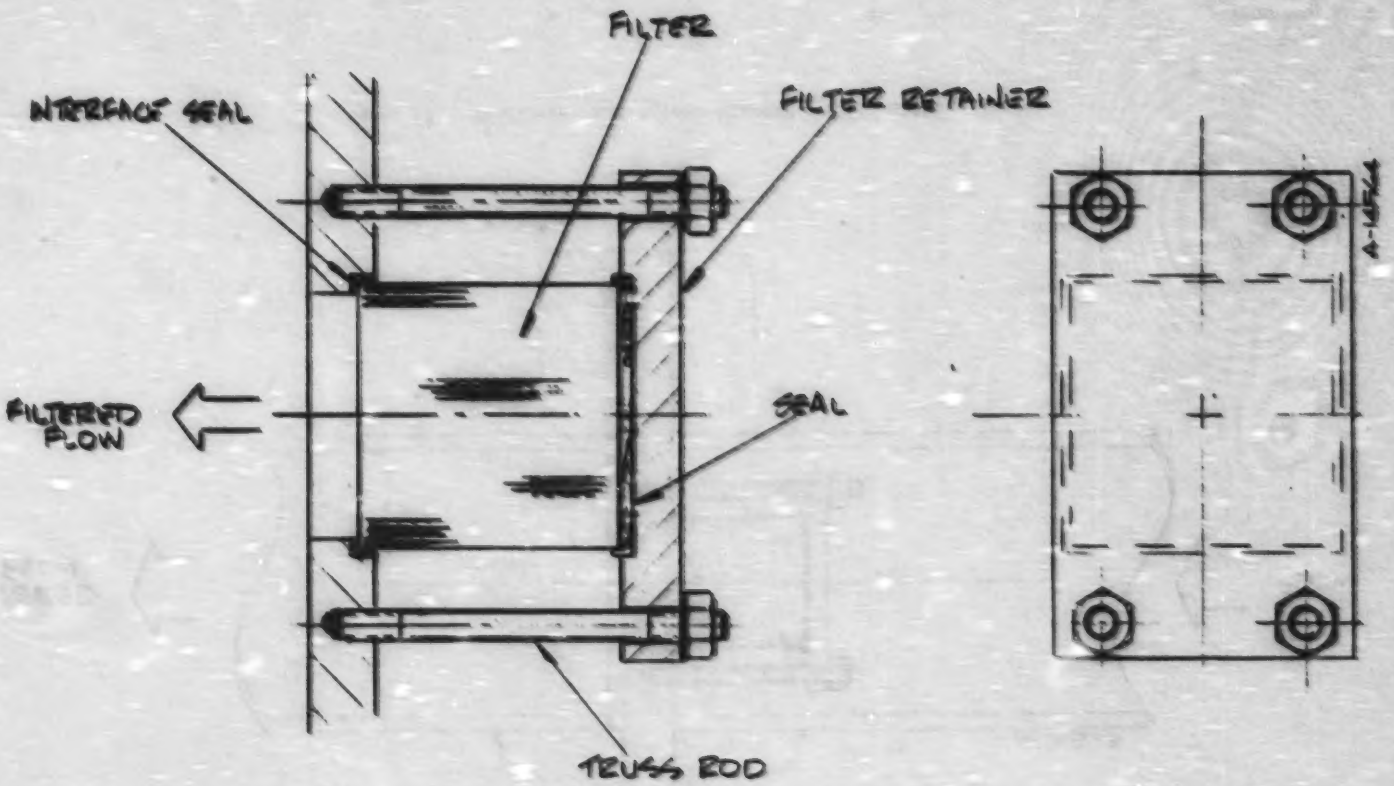


Figure 16. 3M element low temperature holder.

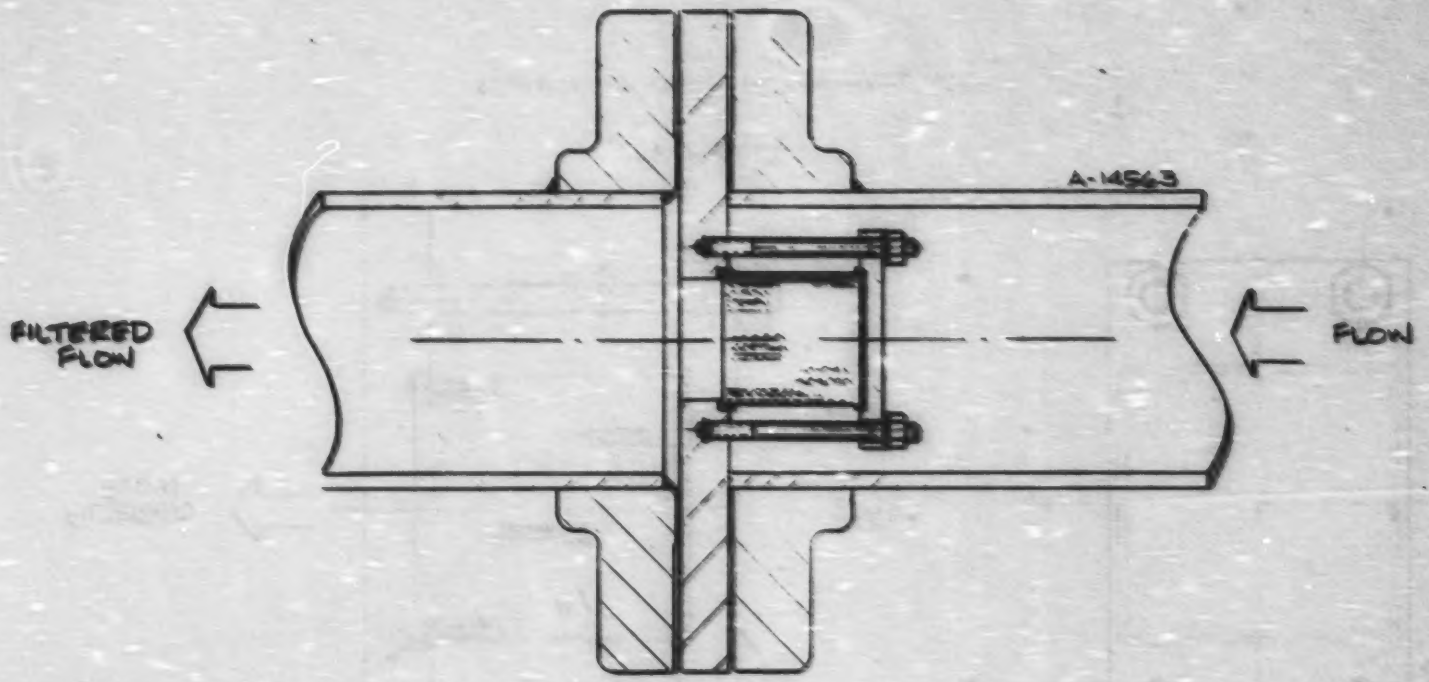


Figure 17. 3M element and holder mounted inside pipe.

A test plan for the evaluation of potential filter materials was prepared and is included as Appendix A. The major test objectives for both low and high temperature testing are:

- Establish pressure drop-flowrate relationships for clean air flow
- Determine fine particle collection efficiency
- Measure pressure drop as a function of time for dirty air
- Investigate various cleaning methods

The testing would be done in two phases. The first phase is for low temperature testing and screening of materials. Only those materials that perform satisfactorily will be tested at high temperatures.

SECTION 7

PROCESS ECONOMICS

Lack of sufficient data at present precludes accurate calculations of process costs for a high temperature particulate cleaning device utilizing ceramic membrane filters. However, sufficient preliminary data do exist to obtain an order of magnitude estimate of the capital requirements and operating costs for a ceramic filter, gas cleaning plant.

Westinghouse Research Laboratories has compiled extensive cost data on ceramic filtration for cleaning fuel gas from a coal gasifier for use in a combined cycle power plant. These data appear in Reference 9 and will only be briefly summarized here.

Westinghouse compared the costs of a ceramic membrane filtration system to those of a combined granular bed/conventional baghouse filtration system. For purposes of illustration, the initial capital investment and the annual operating expenses were estimated for each particulate cleanup system which treats approximately 30 m^3/sec (63,600 ACFM) of fuel gas at 870°C and 1620 kPa (235 psia). This is equivalent to the output of a coal gasifier which supplies fuel to a 250 MW combined cycle plant. An installed capital cost of $8.25 million and total annual operating expenses of $1.8 million were estimated for the granular bed/conventional baghouse system. Estimated costs for the ceramic membrane filter system were made for various filter characteristics - pore diameter, membrane thickness, gas velocity, free cross section, etc. For a filter with 1 micron diameter pores, the estimated capital investment ranged from about $0.7 to $7.0 million, and total annual operating expenses ranged from $0.7 to $2.3 million. For example, for a case requiring 800 m^2* of 0.25 mm thick ceramic membrane filter, the capital investment is $2.1 million and the annual costs are $0.9 million, which is easily competitive with the above alternative.

Several ceramic materials currently produced by ceramic and catalyst monolith suppliers were described in a previous section. Many of these materials have the potential to meet the requirements of a hot gas filtration system. A cost comparison of various filter systems operating at the conditions shown in Table 1 is given in Table 3. These results are for a single

*Pore size = 1μm, superficial gas velocity = 4.5 cm/sec, pressure loss = 60 kPa.

filter system and do not include any duplication which may be required for on-line/off-line operation. The costs shown are for the filter materials only; no pressure vessel or tube sheet costs are included. It is interesting to note that the pressure drop required for either the thick- or thin-walled membranes is significantly lower than the 50 to 80 kPa estimated for reasonable flows through the Westinghouse/Horizons material. This is primarily due to the different pore size and permeability of these materials.

TABLE 3. COMPARISON OF FILTER ELEMENT COSTS FOR THE CONDITIONS IN TABLE 1

	Thick-Wall	Thin-Wall	Westinghouse
Wall Thickness – mm	4.76	0.25	0.25
Flow Area – m^2 (ft^2)	8.17 (88)	24.00 (258)	17.0 (177)
Superficial Velocity – cm/sec (in/sec)	9.1 (3.57)	2.54 (1.0)	4.5 (1.77)
Approximate Pressure Drop, Clean – kPa (psi)	34.5 (5)	3.732 (0.54)	60.0 (8.7)
Size of Elements – mm (in)	66.7 dia x 254 x 4.8 (2-5/8 dia x 10 x 3/16)	152 x 152 x 152 (6 x 6 x 6)	–
Number of Elements	153	17	–
Approximate Cost of Elements	$4600 ($560/m^2)	$850 ($35.50/m^2)	$4080 ($240/m^2)

The most interesting aspect of the cost comparison is the projected cost for thin-wall ceramic material in monolith form. Currently available material at $35.50/$m^2$ for 4 cells/cm is roughly 10 times less expensive than the projected cost of the Westinghouse ceramic material. If, for either material, the costs associated with plant construction are approximately equivalent, and if the element cost is a significant portion of total cost of the system, then the use of ceramic monoliths offers a great potential cost savings.

It appears that the economics of ceramic membrane filters, in particular the ceramic monoliths, compare quite favorably with other gas cleanup schemes for particulate removal from high temperature gas streams.

SECTION 8

CONCLUSIONS AND RECOMMENDATIONS

This investigation resulted in the following conclusions:

- The theory for the operation of membrane filters is not adequate to predict the operation of real filters.
- The Westinghouse project has had only limited success thus far in developing and testing a ceramic membrane filter.
- Current available ceramic filters
  - appear promising for high temperature fine particulate control
  - appear to be economically feasible
  - need to be experimentally tested for high temperature operation.
- Ceramic honeycomb monoliths
  - offer excellent possibilities for use in high temperature particulate control
  - are economically superior to present ceramic filter elements
  - need to be experimentally tested in particulate control applications.

Before adequate assessment of these control methods can be made, several important questions must be answered. These are:

- What is the relationship of collection efficiency to pore size and pore size distribution and to particulate size and size distribution?
- What is the relationship of pressure loss to pore size, porosity, and permeability?
- How susceptible to clogging are the ceramic elements?
- How can the elements be cleaned?

An experimental program that will provide answers to the above questions is recommended. This program utilizes currently available ceramic filter elements and ceramic monoliths, and thus, requires no material development.

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APPENDIX A

FILTER TEST PLAN

1. NOMENCLATURE

A – Surface area available for filtration (cm^2)

B – Inlet gas particle loading (g/cm^3)

P – Pressure (kPa)

ΔP – Pressure drop across a filter element (kPa)

Q – Gas flowrate (cm^3/sec)

t – Time (sec)

T – Temperature (°K)

V_o – Superficial gas velocity, Q/A (cm/sec)

η – Particle collection efficiency

2. AVAILABLE TEST FACILITIES

Several of Westinghouse Research Laboratories' filter test facilities are available for use. The flow conditions for three of them, which are to be used in the initial phase of the testing, are given below.

2.1 10 Liter/min Facility

- Q from 15 to 150 cm^3/sec (1 to 10 ℓ/min)
- Maximum ΔP of 50 kPa (7 psi)
- T to 1150°K (1600°F)
- Atmospheric pressure
- Polydisperse particles, sub-10 μm diameter
- Particle loading limits unspecified (but nominally 0.1 to 0.5 grain/SCF)

2.2 300 Liter/min Facility

- Q from 500 cm^3/sec to 5000 cm^3/sec (30 ℓ/min. to 300 ℓ/min)
- Maximum ΔP of 6 kPa (1 psi)
- Ambient temperature
- Atmospheric pressure
- Polydisperse particles, sub-10 μm diameter
- Particle loading limits unspecified (but nominally 0.1 to 0.5 grain/SCF)

2.3 High Flowrate Facility

- Q up to 2.36 x $10^5 cm^3$/sec (14,158 ℓ/m, 500 acfm at 1500°F)
- Maximum ΔP of 7.5 kPa (7.1 psi)
- Temperatures up to 1150°K (1500°F)
- Atmospheric pressure
- Polydisperse particles, sub-10 μm diameter
- Particle loading limits unspecified (but nominally 0.1 to 0.5 grain/scf)

3. FILTER TEST MATERIALS

Three types of ceramic filter materials will be tested. If other materials become available they will be included in the Phase 1 testing.

3.1 Thick Walled (1/8" to 3/8") Ceramic Filter Disks

Several disks will be tested in the 1 to 10 ℓ/min. facility. Filter properties to be varied include pore size and disk thickness.

3.2 Thin Walled (~0.010") Ceramic Catalyst Monoliths

Several monoliths will be tested in the 30 to 300 ℓ/min. facility. The cell size and wall thickness may be varied.

3.3 Alumina FiberForm

Alumina FiberForm, manufactured by Fiber Materials, Inc., is a new material made of alumina-silica fibers bonded with a high purity alumina binder and produced using vacuum slurry molding techniques. This material is significantly different from the ceramic filter materials and the

ceramic monoliths. It has a useful temperature unit of about 1450°C (2600°F) and a very high porosity.

4. PRETEST ACTIVITY

- Westinghouse will procure suitable ceramic filter disks and disk holders
- Aerotherm will procure catalyst monolith materials and FiberForm, and deliver them to Westinghouse

5. FILTER TEST MATRIX

Filter testing will proceed in two phases - Phase 1 for ambient temperature tests and Phase 2 for high temperature tests. The Phase 1 testing is intended to screen out materials which are not suitable for use as fine particulate filters.

5.1 Phase 1 - Ambient Temperature Testing (~80°F)

This test phase has the following general objectives:

- Obtain ΔP versus V_o relationships for clean gas flow at ambient temperature
- Evaluate η as a function of t, V_o, and particle size at ambient temperature
- Evaluate the effectiveness of various backflow filter cleaning methods
- Evaluate various filter cycle possibilities

Testing to fulfill these objectives will be accomplished in four test series.

5.1.1 Test Series 1 - Clean Flow Characterization

- Objective:
  - Establish the ΔP versus V_o relationships as functions of filter pore size and thickness for clean gas flow at ambient temperature
- Test Variables:
  - Q - vary over appropriate facility test range
  - Pore size - vary by testing several elements of each type
  - Thickness - vary by testing several elements of each type
- Measure:
  - ΔP as a function of Q

Results from this series will consist of plots of ΔP vs V_o, with filter pore size and thickness as possible parameters.

5.1.2 Test Series 2 – Particulate Removal Efficiency Characterization

- Objectives:
  - Determine ΔP versus t as a function of C, V_o, and filter pore size at ambient temperature.
  - Determine filter collection efficiency (η) versus particle size as a function of C, V_o, and filter pore size
  - Investigate the effects of cake buildup and compression on η.
- Test Variables:
  - Q - vary over appropriate facility test range
  - C - vary in the range 0.2 to 1.0 x 10^{-3} g/cm^3 (0.1 to 0.5 grain/SCF) for each flowrate
  - Pore size - vary by testing several elements of each type
- Measure:
  - ΔP vs t
  - Inlet particle loading and size distribution
  - Outlet particle loading and size distribution as a function of time

Inlet gas particulate load will consist of a polydisperse sub-10μm particle mixture. Each filter element is to be tested at several gas flowrates and several inlet particle loadings. Measurements of ΔP versus t will give the rate of dust cake accumulation on the filter. The influences of gas superficial velocity, inlet particle loading, and filter pore size on this rate of accumulation will be investigated. Comparison of outlet particle load and size distribution with inlet load and distribution will allow calculation of particle collection efficiency versus particle size. The influence of V_o, C, and filter pore size on η will also be investigated. Measurement of η versus t will allow assessment of the relative contributions to particle collection from filter collection and dust cake collection mechanisms. Obviously, the filter element should be thoroughly cleaned between individual experiments. Results from this series will include plots of ΔP versus t, and η as a function of particle size versus t, with C, V_o, and pore size as parameters. The possibility of combining this series with test series 3 should be considered.

5.1.3 Test Series 3 – Cleaning Method Study

- Objective:
  - Investigate various backflow filter cleaning methods including steady backflow and pulsed backflow
- Test Variables:
  - Backflow Q and duration of backflow for steady backflow methods
  - Pulse pressure and duration for pulsed backflow methods
- Measure:
  - Initial ΔP after the cleaning operation

The primary purpose of this study is to observe dust cake removal and pore cleaning. Two types of backflow cleaning experiments should be performed: steady backflow, in which the rate and duration of backflow are varied, and pulsed backflow, where the backflow pressure pulse magnitude and duration are varied. Measurement of the initial, clean gas ΔP (forward flow) will serve as a reference point to define the degree of dust cake removal achieved. Assessment of the degree of short term (one cycle) pore clogging and the effect of V_0 on pore clogging will also be possible. This series will result in the determination of the "best" backflow cleaning methods for each filter element type. The possibility of combining experiments in this series with those in Series 2 should be considered.

5.1.4 Test Series 4 – Cleaning Cycle Study

- Objectives:
  - Determine feasible, long-term filter cycles, including filter times and cleaning times for the "best" cleaning methods from Series 3
  - Investigate the effects of filter pore clogging on long term filter operations, including an estimate of filter lifetime at ambient temperature
  - Investigate the effects of flow channel clogging in monolith elements on long-term filter operation
- Test Variables:
  - Filter time and cleaning time in a complete filter cycle
  - Q
  - C

- Measure
  - ΔP versus t during filtering operation
  - η as a function of particle size

This test series will extend the work of Series 3 by investigating feasible filtering cycles, using the "best" backflow filter cleaning method(s) defined in Series 3. A "best" combination of filtering time, followed by cleaning time, over many filter cycles will be described for long-term filter operations. Since the rate of dust cake accumulation is dependent on filtering flowrate and inlet particulate loading, these variables will be parameters in the cleaning cycle description. As such, Q and C will be varied during this test series. Measurements of ΔP versus t and initial filter cycle ΔP will be used to evaluate candidate filter cycles. Occasional measurements of particulate collection efficiency should also be performed to monitor filter performance. It is expected that initial cycle ΔP will increase with time as filter pores clog; therefore, the length of the filtering portion of a cycle will have to decrease as number of cycles performed increases. The rate of this increase in initial ΔP with time will serve to define the useful life of a filter element. An additional concern relates to flow channel clogging in monolith elements. Backflow alone may not completely clear these channels. The effects of this clogging on element usable lifetime may also be assessed by noting the time rate of increase in initial cycle ΔP. Results from this series will consist of time specifications for "best" feasible filter-clean cycles, together with estimates of filter element usable life.

5.2 Phase II – High Temperature Testing

High temperature testing will be performed for those materials identified in the Phase I testing as potential filters. General objectives for this phase of testing include:

- Obtain ΔP versus V_o relationships for clean gas as a function of temperature
- Assess potential filter porosity decreases, due to material phase change or material sintering, on ΔP vs V_o relationships
- Evaluate η as a function of particle size and temperature at constant V_o and C
- Evaluate candidate filter-cleaning cycles at high temperatures

Testing to fulfill these objectives at temperatures up to 1100°K (1500°F) will be accomplished in four additional test series.

5.2.1 Test Series 5 — High Temperature Clean Flow Characterization

- Objectives:
  - Determine ΔP vs V_o for clean gas flow as a function of temperature
  - Determine if changes in the ΔP, V_o curves can be accounted for solely by gas viscosity changes or are material pore structure changes also important
- Test Variables:
  - T - vary temperature to 1100°K (1500°F)
  - Q - vary flowrate over appropriate facility test range
  - Pore size - vary by testing several elements of each type
- Measure:
  - ΔP as a function of Q at each T

ΔP is expected to increase with increasing temperatures, at constant V_o, due to

- Increasing gas viscosity with increasing T
- Potential porosity decreases due to phase change, or material sintering

Results of this series will consist of plots of ΔP versus V_o with temperature as a parameter for each pore size tested, and a determination of the relative effects of gas viscosity and decreases in porosity on ΔP-V_o curves.

5.2.2 Test Series 6 — High Temperature Particulate Removal Efficiency Characterization

- Objectives:
  - Investigate ΔP versus t as a function of filter pore size and temperature
  - Investigate η as a function of filter pore size and temperature
- Test Variables:
  - T - vary to 1100°K (1500°F)
  - Pore size — vary by testing several elements of each type
- Measure:
  - ΔP versus t
  - Inlet particle size distribution
  - Outlet particle loading and size distribution as a function of time

The test series is essentially a repeat of Series 2 with the introduction of temperature as a primary test variable. The theory of filter particle collection mechanisms predicts that η should decrease with increasing temperature. This would imply that the rate of dust cake accumulation

(and the slope of a ΔP versus t plot) should be smaller at high temperatures. However, these effects may be offset by potential porosity decreases as discussed in Section 5.2.1. The findings of this test series, taken with those of Series 5, will serve to define the relative magnitude of the effects of possible decreased porosity versus decreased collection efficiency on overall filter collection at high temperature. Results from this series will consist of plots of ΔP versus t, with T as a parameter, at varying pore size, and η as a function of particle size versus T at varying filter pore size.

5.2.3 Test Series 7 – High Temperature Cleaning Cycle Study

- Objective:
  - Determine feasible high temperature cleaning cycles
- Test Variables:
  - Filter time, cleaning time
- Measure:
  - ΔP versus t during filtering operation
  - η as a function of particle size

This series is essentially a high temperature repeat of Test Series 4. In this series, testing at one flowrate, one particle loading, and one temperature should suffice. Test temperature should be the highest temperature expected in eventual ceramic filtration applications (~1500°F). The purpose of this test series is to refine and extend the conclusions derived in Test Series 4 to high temperature. Results from this test series will consist of the time specifications for "best" feasible high temperature filter-clean cycles, together with estimates of filter element usable life at high temperature.

5.2.4 Test Series 8 – High temperature, High Flow Tests

This test series will be conducted only with those materials that perform satisfactorily in Test Series 5 through 7. The objective of this test is to simulate as close as possible "real world conditions. These tests will be done in the high flow facility described in Section 2.3. The test will encompass the activities described in Test Series 6 and 7.